## THE POPOP4 LIBRARY AND CODES FOR PREPARING SECONDARY GAMMA-RAY PRODUCTION CROSS SECTIONS

## W. E. Ford, III

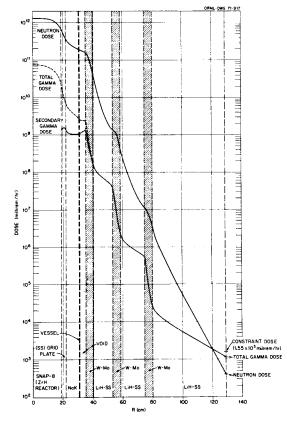
## Oak Ridge National Laboratory Oak Ridge, Tennessee

The POPOP4 code for converting secondary gamma-ray yield data to multigroup secondary gamma-ray production cross sections and the POPOP4 library of secondary gamma-ray yield data are described. Recent results in the "testing" of uranium and iron data sets from the POPOP4 library are given. The data sets were tested by comparing calculated secondary gamma-ray pulse-height spectra with spectra measured at the ORNL TSR-II reactor.

One of the primary considerations in the design of shields for reactors on spacecraft is the dose rate due to secondary gamma rays produced by neutron interactions in the fuel and in the structural and shielding materials. The importance of secondary gamma rays is illustrated by the calculated dose distribution shown in figure 1 for a three-cycle W-LiH shield for a SNAP-8 ZrH reference reactor (ref. 1). The design objectives were to limit the dose rate on the outer surface of the shield to a maximum value of  $1.55 \times 10^3$  mrem/hr and to minimize the weight of the spherical shield. Note from the figure that before the constraint dose is obtained, the secondary gamma-ray dose rate predominates. These dose rates were calculated by coupling the transport of the neutrons and secondary gamma rays in a single discrete ordinates calculation. One of the requirements for a coupled calculation is accurate secondary gamma-ray production cross sections (SGRPXS's). To satisfy this requirement, the shield designer must have a readily accessible library of secondary gamma-ray yield data for the elements used in shield design, the means to convert the yield data to SGRPXS's in a format for use with the standard transport codes, and confidence in the accuracy of the data.

POPOP4, a FORTRAN-IV code, was written to convert secondary gamma-ray yield data as found in the literature for (n,γ), (n,xγ), etc., reactions to any required neutron-gamma multigroup energy structure (ref. 2). If the yield data are in terms of gamma-ray intensity per neutron induced reactions, the code multiplies the converted multigroup yields by input multigroup neutron-reaction cross sections to give the SGRPXS's. [The neutron reaction cross sections are obtained from codes such as GAM-II or XSDRN (refs. 3 and 4).] POPOP4 sums the SGRPXS's for the various neutron-induced reactions to give

total SGRPXS's for the nuclide of interest. Using codes such as the Sample Simple Coupling Code (ASSCC) (ref. 5), the POPOP4 cross sections are coupled with  $P_N$  neutron and gamma-ray cross-section sets for use in coupled neutron-gamma transport calculations. Discrete ordinates codes such as ANISN (ref. 6) and DOT (ref. 7) and Monte Carlo codes such as MORSE (ref. 8) use multigroup coupled cross sections produced as described above.



Dose Distribution in Snap-8 Reactor with Three-Cycle W-LiH Shield,

FIGURE 1.-Dose Distribution in SNAP-8 Reactor With Three-Cycle W-LiH Shield.

A compendium of neutron-induced secondary gamma-ray yield and cross-section data has been compiled for use with POPOP4. This collection of data is known as the POPOP4 library (ref. 9). Included in the library are capture and inelasticscattering yield data sets from the United Nuclear Corporation publications (refs. 10, 11, and 12), Maerker's and Muckenthaler's measured secondary gamma-ray yields due to thermal-neutron captures in the elements found in soils, concretes, and structural materials (ref. 13), the yields for thermalneutron captures in a large number of elements as compiled in the Nuclear Data publications (refs. 14, 15, and 16), the isotropic components of the gammaray production cross sections for Na, Mg, Cl, K, and Ca reported by Drake  $et\ al.$  (ref. 17), and many other  $(n,\gamma)$  and  $(n,x\gamma)$  data sets from the literature and from the private files of contributors. At present there are 223 data sets in the library for 79 elements or nuclides. The library is available on magnetic tape from the Radiation Shielding Information Center (RSIC). [POPOP4, the Sample Simple Coupling Code, and POPOP4 Library Tape Maker (ref. 5) - a code to make or update the POPOP4 library tape - are also available from RSIC.]

One of the most important objectives of the POPOP4 project is the "testing" of secondary gamma-ray yield data for use in shielding calculations. The data are tested by comparing measured secondary gamma-ray pulse-height spectra with spectra calculated using POPOP4 multigroup SGRPXS's prepared with data sets from the POPOP4 library.

A series of experiments was conducted by Muckenthaler et al. of ORNL's Neutron Physics Division to measure secondary gamma-ray pulse-height spectra resulting from the exposure of various slabs of reactor core, structural, and shielding materials to collimated neutron beams (refs. 13 and 18). The slabs were exposed to bare, Cd-filtered, and 10Bfiltered neutron beams emanating from the ORNL TSR-II reactor. A simplified illustration of the experimental configuration is shown in figure 2. Differential pulse-height spectra due to secondary gamma rays produced by neutron-induced reactions in the slabs were measured at a point 20 ft from the slabs. A borated polyethylene filter was placed between the NaI(Tl) detector and the slab to reduce thermal-neutron effects in the detector. Some materials

required an additional LiH filter in front of the borated polyethylene to thermalize scattered neutrons. These experiments provided the standards for testing the data sets from the POPOP4 library.

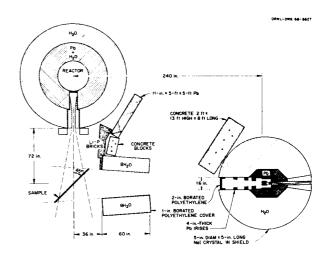


FIGURE 2.-TSR-II Experimental Configuration for Measuring Secondary Gamma-Ray Pulse-Height Spectra.

A flowchart illustrating the general procedure for calculating secondary gamma-ray pulse-height spectra is shown in figure 3. SGRPXS's were prepared by using POPOP4 to convert the yield data sets being tested to a 27 neutron - 60 gamma energy group structure and, if the data were given in terms of secondary gamma-ray yields, to combine the data with spectrally weighted, 27 group neutron reaction cross sections calculated with XSDRN or GAM-II. ASSCC was used to couple the SGRPXS's, P3 neutron cross sections, and P3 MUG (ref. 19) gamma-ray cross sections for the element of interest. Using multigroup representations of the TSR-II beams and the coupled cross sections as input, the one-dimensional discrete ordinates transport code ANISN was used to calculate the secondary gamma-ray angular flux emanating from the slabs. All ANISN calculations were made using a four-term expansion of Legendre polynomials and a 96th order of angular quadrature.

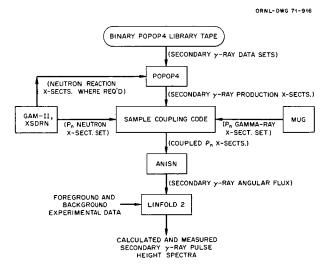


FIGURE 3.-Calculational Procedure for 'Testing' Secondary Gamma-Ray Yield Data.

Calculational Procedure for "Testing" Secondary Gamma-Ray Yield Data

The FORTRAN-IV code LINFOLD (ref. 5) was used to correct the ANISN angular flux for the geometric attenuation and for the attenuating effect of the detector neutron shield(s) - giving the intensity of secondary gamma rays striking the detector within each gamma group. One-fifth of the intensity within each group was assumed to be the intensity of five equally spaced discrete gamma energies within the group. Using Maerker's experimentally determined response functions for the NaI(Tl) detection system (ref. 13), the intensities of the discrete gammas were folded with LINFOLD to give the calculated spectra. Plots of the calculated and measured spectra were produced as the means for comparison.

Initial efforts in the data testing program were concentrated on the 23 Al, Fe, Ni, Cu, and Pb  $(n,\gamma)$  and  $(n,n'\gamma)$  data sets from refs. 11, 12, 13, 14, and 16. Pictorial comparisons of the calculated and measured pulse-height spectra resulting from the exposure of slabs of these materials to the TSR-II neutron beams are shown in ref. 20. The comparisons provide a means of evaluating the validity of the data sets for various incident neutron energy ranges. Recent efforts have been directed to the testing of the U, W, and Ta data sets in the library and to the testing of a newly acquired Fe  $(n,\gamma)$  data set. To illustrate the testing procedure, recent results for U and Fe are described below.

Muckenthaler et al. measured the spectra resulting from the exposure of a 10.44, x 5.75 x 0.0304in.-thick depleted uranium foil to the collimated TSR-II beams. Background was determined for each beam by measuring the spectra without the foil in place. The  $^{235}$ U and  $^{238}$ U number densities in the sample were  $8.305 \times 10^{-5}$  and  $5.02 \times 10^{-2}$  nuclei/ barn cm, respectively. The foil was oriented as shown in figure 2 above. Since gamma rays are produced by the fission capture, non-fission capture, and the inelastic-scattering reactions in both 235U and 238U, and since the SGRPXS's depend on the resonance characteristics of the nuclides, the spectra comparisons for the foil provided an interesting challenge. XSDRN was used to calculate 235U and  $^{238}$ U  $P_3$  neutron cross sections for the 27 neutron group structure. This calculation included the resonance self-shielding effects in both nuclides. Booth has shown that the secondary gamma-ray spectrum resulting from exposure of the foil to the Cd-filtered neutron beam minus 1.377 times the 10B-filtered spectrum eliminates the effect of high-energy (fission) neutrons (ref. 21). Sample comparisons for the (Cd-1.377  $^{10}$ B) and bare neutron beams are shown in figure 4. The data sets used in the preparation of the SGRPXS's for the transport calculations are listed in Table I. Calculation B is a bare beam calculation which included SGRPXS's for all 235U and <sup>238</sup>U neutron induced reactions except the  $^{238}$ U (n, $\gamma$ ) reaction. SGRPXS's for all 235U and 238U neutron induced reactions were used in calculation D. The <sup>238</sup>U SGRPXS's for calculation D were prepared from data sets 928112, 928301, 928901, 925801, and 925804 as indicated in Table I. Calculation A is a (Cd-1.377  $^{10}$ B) calculation which included SGRPXS's for all reactions except the  $^{238}U$  (n, $\gamma$ ) reaction, whereas, calculation C included SGRPXS's for all 235U and <sup>238</sup>U reactions. A report describing similar tests of twelve uranium data sets from the POPOP4 library is in the final stages of preparation (ref. 25).

Measured spectra resulting from the exposure of an approximately 5-ft-square by 1/16-in.-thick Fe slab to the TSR-II beams were used as the standards for testing Fe data sets. Since the Cd-filter has a "cutoff" at 0.5 eV and a constant attenuation of 0.986 to neutrons above 1 MeV, and since the boron filter "cuts off" at approximately 10 keV and has

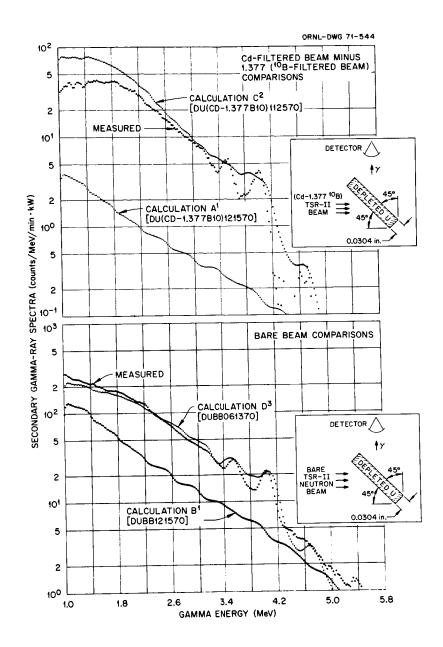


FIGURE 4.-Comparisons of Calculated and Measured Secondary Gamma-Ray Pulse-Height Spectra for a Depleted Uranium Foil.

<sup>\*</sup>Ordinate caption should read Secondary Gamma-Ray Pulse-Height Spectra (counts/MeV/min·kW).

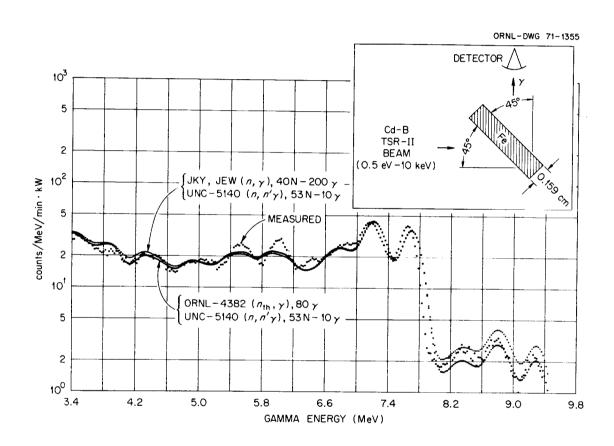


FIGURE 5.-Comparisons of Calculated and Measured Secondary Gamma-Ray Pulse-Height Spectra For an Iron Slab.

Table I. Uranium Secondary Gamma-Ray Yield Data

Data Set Identification	Uranium Isotope/	Source of		
Number <sup>†</sup>	Reaction	Yield Data	Remarks	
925101	5/(n,y)*	Ref. 12, p. 45	Yields given for 11 neutron groups $(3.7 \times 10^{-2} \le E_n \le 18.2 \times 10^6 \text{ eV})$ and 13 discrete gamma energies $(0.0894 \le E_\gamma \le 6.42 \text{ MeV})$ . Data used for the $^{2.35}\text{U}(n,\gamma)$ reaction in all calculated spectra shown in Fig. 4.	
925301	5/(n,n')	Ref. 12, p. 52	Yields given for 80 neutron groups $(1.27 \times 10^4 \le E_n \le 18.017 \times 10^6$ eV) and 11 gamma groups $(0.0 \le E_{\gamma} \le 6.50$ MeV). Data used for the $^{2\$5}$ U(n,n') and (n,2n') reactions in all calculated spectra shown in Fig. 4. Assumed applicable to $^{2\$8}$ U in Calculations A, B, and C.	
925801	5/(n,γ) <sub>F</sub> * and (n,2n')	Ref. 22	Thermal-neutron prompt and delayed fission capture yields given for 20 gamma groups $(0.0 \le E_{\gamma} \le 6.50 \text{ MeV})$ . These data assumed applicable to both $^{235}\text{U}$ and $^{238}\text{U}$ and to all incident neutron energies in Calculations A, B, and C. Data used in Calculation D for all neutron energies in $^{235}\text{U}$ SGRPXS and for $E_{n} \le 1.1$ MeV in $^{238}\text{U}$ SGRPXS.	
925804	5/(n,γ) <sub>F</sub>	Ref. 22	Delayed part of 925801 data (t > 1 sec). Data used only in Calculation D for $\rm E_n$ > 1.1 MeV in $^{238}\rm U$ SGRPXS.	
928113	8/(n,y) <sub>NF</sub>	Ref. 23, pp. 51-65	Yields given for 21 neutron groups $(5.6 \le E_n \le 1.019 \times 10^5 \text{ eV})$ and 16 gamma groups $(0.903 \le E_\gamma \le 4.807 \text{ MeV})$ . Data used in Calculation C for $^{238}\text{U}(n,\gamma)$ SGRPXS in applicable neutron energy range.	
92811 <b>2</b>	8/(n, y) <sub>NF</sub>	Ref. 26	Yields given for 37 neutron groups (0.0 $\leq$ E $_n$ $\leq$ 11.08 MeV) and 117 gamma groups (0.0 $\leq$ E $_{\gamma}$ $\leq$ 5.85 MeV). Data used in Calculation C for E $_n$ < 5.6 eV and E $_n$ > 1.019 x 10 $^5$ eV. Data used in Calculation D for E $_n$ < 1.1 MeV.	
928301	8/(n,n') and (n,2n')	Ref. 12, p. 54	Yields given for 79 neutron groups (0.337 $\leq$ E <sub>n</sub> $\leq$ 18.02 MeV) and 11 gamma groups (0.25 $\leq$ E $_{\gamma}$ $\leq$ 6.5 MeV). Data used in Calculation D for E <sub>n</sub> $<$ 1.1 MeV.	
928901	8/(n,y) <sub>NF</sub> , (n,y) <sub>F</sub> , and (n,n')	Ref. 24, p. 19	SGRPXS given for 15 neutron groups (0.485 $\leq$ E <sub>n</sub> $\leq$ 15.3 MeV) and 12 gamma groups (0.50 $\leq$ E <sub><math>\gamma</math></sub> $\leq$ 6.5 MeV). X-sects used in Calculation D for <sup>238</sup> U(n, $\gamma$ ) <sub>NF</sub> , (n, $\gamma$ ) <sub>prompt</sub> F, and (n,n') reactions in applicable neutron energy range.	

 $<sup>*(</sup>n,\gamma)_{NF}$  is the nonfission capture reaction and  $(n,\gamma)_F$  is the fission capture reaction.

Table II. Iron Secondary Gamma-Ray Yield Data

Data Set Identification Number	Reaction	Source of Yield Data	Remarks
260105	(n,γ)	Ref. 13, p. 67	Thermal-neutron capture yields given for 80 discrete gamma energies (1.24 $\leq$ $E_{\gamma}$ $\leq$ 9.30 MeV). These data assumed applicable to all incident neutron energies in calculation labeled "ORNL-4382"
260110	(n,γ)	Ref. 26	Yields given for 40 neutron groups $(0.0 \le E_n \le 1.0026$ MeV) and 200 gamma groups $(0.0 \le E_{\gamma} \le 10.0$ MeV). Data used in calculation labeled "JKY, JEW"
260301	(n,n')	Ref. 12, p. 35	Yields given for 53 neutron groups (0.84 $\leq$ E <sub>n</sub> $\leq$ 18.1 MeV) and 10 gamma groups (0.50 $\leq$ E $_{\gamma}$ $\leq$ 10.0 MeV). Data used in both calculations shown in Fig. 5.

 $<sup>^\</sup>dagger$ Each data set in the POPOP4 library is identified by a unique six-digit identification number.

a constant attenuation of 0.75 to neutrons above 1 MeV. the spectra from the (Cd-10B) neutron beam provide a test of the epithermal Fe capture yield data (ref. 18). Two calculated spectra resulting from exposure of the slab to the (Cd-10B) beam are compared with the measured spectrum in figure 5. The data sets used in the preparation of SGRPXS for the calculations are identified in Table II. Neutron cross sections for the calculations were calculated with GAM-II.

Results of these studies have proven that the conversion technique used in POPOP4 is satisfactory for the conversion of yield data to a required multigroup energy structure. Since the SGRPXS's are only as accurate as the yield data and the neutron reaction cross sections, the neutron cross sections must be properly weighted prior to being used. The growth envisioned for the POPOP4 library depends on the continued support of individuals who are willing to share their efforts with others.

## REFERENCES

- 1. The calculation shown in figure 1 was made by R. L. Childs of Oak Ridge National Laboratory.
- 2. FORD, W. E., III; and WALLACE, D. H.: POPOP4 A Code for Converting Gamma-Ray Spectra to Secondary Gamma-Ray Production Cross Sections. USAEC Report CTC-12, Union Carbide Corporation, 1969.
- 3. JOANOU, G. D.; and DUDAK, J. S.: GAM-II, A  $B_3$  Code for the Calculation of Fast-Neutron Spectra and Associated Multigroup Constants. Gulf General Atomic Report GA-4265, 1963.
- 4. GREENE, N. M.; and CRAVEN, C. W., JR.: XSDRN A Discrete Ordinates Spectral Averaging Code. USAEC Report ORNL-TM-2500, Union Carbide Corporation, 1969.
- 5. The Sample Simple Coupling Code, POPOP4 Library Tape Maker, and LINFOLD are documented in reference 20.
- 6. ENGLE, W. W.: A User's Manual for ANISN A One-Dimensional Discrete Ordinates Transport Code With Anisotropic Scattering. USAEC Report K-1693, 1967.
- 7. MYNATT, F. R.: A User's Manual for DOT. USAEC Report K-1694 (to be published).
- 8. STRAKER, E. A.; STEVENS, P. N.; IRVING, D. C.; and CAIN, V. R.: The MORSE Code - A Multigroup Neutron and Gamma-Ray Monte Carlo Transport Code. USAEC Report ORNL-4585, 1970.
- 9. FORD, W. E., III: The POPOP4 Library of Neutron-Induced Secondary Gamma-
- Ray Yield and Cross Section Data. USAEC Report CTC-42, 1970. 10. TROUBETZKOY, E. S., et al.: Neutron Cross Sections of  $U^{238}$ ,  $U^{235}$ ,  $U^{237}$ ,  $U^{239}$ ,  $U^{234}$ ,  $U^{236}$ ,  $Pu^{239}$ ,  $Pu^{240}$ , W, Pb, Ni, Cr, C, Li<sup>6</sup>, Li<sup>7</sup>, and T. United Nuclear Corporation Report UNC-5099, 1964.
- 11. RAY, J. H.; TROUBETZKOY, E. S.; and GROCHOWSKI, G.: Neutron Cross Sections of Nitrogen, Oxygen, Aluminum, Silicon, Iron, Deuterium, and Beryllium. United Nuclear Corporation Report UNC-5139, 1965.
- 12. CELNIK, J.; and SPIELBERG, D.: Gamma Spectral Data for Shielding and Heating Calculations. NASA Report NASA CR-54794 (UNC-5140), 1965.
- 13. MAERKER, R. E.; and MUCKENTHALER, F. J.: Gamma-Ray Spectra Arising From Thermal Neutron Capture in Elements Found in Soils, Concretes, and Structural Materials. USAEC Report ORNL-4382, 1969.
- 14. WAY, K., Ed.: Compendium of Thermal-Neutron-Capture Gamma-Ray Measurements, Part I. Z ≤ 46. Nuclear Data, vol. III, nos. 4-6, pp. 367-650, 1967.
- 15. WAY, K., Ed.: Compendium of Thermal-Neutron-Capture Gamma-Ray Measurements, Part II. Z = 47 to Z = 67 (Ag to Ho). Nuclear Data, vol. V, nos. 1-2, pp. 1-242, 1968.
- 16. WAY, K., Ed.: Compendium of Thermal-Neutron-Capture Gamma-Ray Measurements, Part III. Z = 68 to Z = 94 (Er to Pu). Nuclear Data, vol. V, nos. 3-4, pp. 243-431, 1969.
- 17. DRAKE, M. K.; GARRISON, J. D.; and ALLEN, M. S.: Neutron and Gamma Ray Production Cross Sections for Sodium, Magnesium, Chlorine, Potassium, and Calcium. Nuclear Defense Laboratory Reports NDL-TR-89, vols. I-VI, 1967.

- MAERKER, R. E.; and MUCKENTHALER, F. J.: Gamma-Ray Spectra Arising From Fast-Neutron Interactions in Elements Found in Soils, Concretes, and Structural Materials. USAEC Report ORNL-4475, 1970.
- KNIGHT, J. R.; and MYNATT, F. R.: MUG A Program for Generating Multigroup Photon Cross Sections. USAEC Report CTC-17, 1970.
- 20. FORD, W. E., III; and WALLACE, D. H.: The Use and 'Testing' of A1, Fe, Ni, Cu, and Pb Secondary Gamma-Ray Production Data Sets From the POPOP4 Library. USAEC Report CTC-20, 1970.
- Library. USAEC Report CTC-20, 1970.

  21. BOOTH, R. S.; and MUCKENTHALER, F. J.: The Total Gamma-Ray Energy Spectrum Above 1 MeV From Neutron Interactions in 235U and 238U. USAEC Report ORNL-TM-3060, Union Carbide Corporation, 1971 (to be published).
- 22. CLAIBORNE, H. C.: private communication.
- JOHN, J.; and ORPHAN, V. J.: Gamma Rays From Resonant Capture of Neutrons in <sup>238</sup>U. Gulf General Atomic Report GA-10186, 1970.
- NELLIS, D. O.; and MORGAN, I. L.: Gamma-Ray Production Cross Sections for 235<sub>U</sub>, 238<sub>U</sub>, and 239<sub>Pu</sub>. Texas Nuclear Corporation Report ORO-2791-17, 1966.
- 25. FORD, W. E., III: The Testing of Uranium Secondary Gamma-Ray Production Data Sets From the POPOP4 Library - the 30-Mil Foil Comparisons. USAEC report (to be published).
- 26. YOST, K. J.; et al.: private communication.